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(54) THERMALLY WELDABLE LACTIC ACID POLYMER LAMINATE

## (57) Abstract:

**PROBLEM TO BE SOLVED:** To provide a good heat resistivity, sealing strength, and a biodegradation property by having a foundation layer consisting of a crystallized lactic acid polymer and a seal layer consisting of noncrystal lactic polymer having a lower melting point than the lactic polymer.

**SOLUTION:** Thermally weldable lactic acid polymer laminate has a bare layer I consisting of crystallized lactic polymer A being a layer of realizing a heat resistive or heat molding property and a seal layer II consisting of a noncrystal lactic acid polymer B to be thermally welded by a beat sealing method or the like and having a lower melting point of the lactic acid polymer A. The crystallized lactic acid polymer A is such that a polyester constitutional unit being dehydrated-and-condensed with dicarbonic acid and diol and/or a 3-60wt.% polyether constitutional unit being dehydrated-and-condensed with dicarbonic acid and polyether polyol are contained in a constitutional unit being dehydrated-and-condensed with polylactic acid or lactic acid at a melting point of 120°C or higher.

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**DETAILED DESCRIPTION**

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**[Detailed Description of the Invention]****[0001]**

**[Field of the Invention]** This invention relates to containers, such as a package bag which comes to carry out thermoforming of the lactic-acid system polymer layered product and this layered product suitable for a package or receipt of various kinds of food to which it comes to carry out thermal melting arrival of the lactic-acid system polymer layered product excellent in thermal melting arrival nature, thermal resistance, and biodegradability, and this layered product, a drink, a chemical, miscellaneous goods, etc. or a case, and a lightweight container.

**[0002]**

**[Description of the Prior Art]** Although the amount with huge plastics is used in recent years, the trash has caused terrestrial environmental problems, such as a threat to the lack of reclaimed ground, scene inhibition, and a marine organism, and environmental pollution. The plastics called general-purpose resin currently generally used conventionally is polyethylene, polypropylene, polystyrene, a polyvinyl chloride, polyethylene terephthalate, etc., and incineration and reclamation are performed as the disposal approach of these resin.

[0003] However, since the combustion calorie of these resin is high when there is a problem in these disposal approaches and it incinerates resin, such as polyethylene, polypropylene, and polystyrene, it is easy to damage a furnace and the life of a furnace is shortened. On the other hand, although a combustion calorie is low in a polyvinyl chloride, generating gas harmful at the time of incineration is known. Also in reclamation, since these general-purpose resin has high chemical stability, remaining semipermanently, not decomposing but stopping the original form is known, and it has become one of the causes which the lack of reclaimed ground aggravates.

[0004] Moreover, when easily discarded in natural environment, for the stability, a fine sight is spoiled, or a marine organism, birds, etc. carry out an adding diet accidentally, and it has become the cause of environmental destruction -- a precious biomass decreases. In order to solve these problems, research of a biodegradability polymer is done briskly recently. The polylactic acid called a lactic-acid system polymer and its copolymer are in one of the resin which attracts attention by the biodegradability polymer. Unlike general plastics, full decomposition of these lactic-acid system polymers is carried out easily, and, finally they become water and a carbon dioxide.

[0005] Moreover, since the combustion calorie is low, also when it destroys by fire, it has the description which does not damage a furnace and does not generate the still more harmful gas at the time of combustion. a start raw material -- playback -- since an easy biomass can be used, it can be freed from drained petroleum resources. From these advantages, it is expected as an alternative of general-purpose resin.

[0006] Conventionally, the film into which paper and synthetic resin were processed, a sheet or aluminum foil, etc. is used as a package of the liquefied objects various kinds of food, a drink, a chemical, for miscellaneous goods, etc., a powdery and granular material, and a solid, or receipt material. the bag aiming at a package or receipt since especially a film and a sheet have the description

excellent in \*\*, such as a water resisting property, transparency, reinforcement, thermoforming nature, and low cost nature, and a case -- or thermoforming is carried out and it is used for many applications as a lightweight container. Thermal melting arrival nature and thermal resistance are mentioned to the important thing as demand characteristics to these packages or receipt material.

[0007] The film and sheet which consist of synthetic resin are processed into various bags or a case by using the thermal melting arrival nature of resin and pasting up one or more ways, bending. Moreover, a film and a sheet are fabricated by the lightweight container which has rigidity and packs contents, such as food, a drink, a chemical, and miscellaneous goods, by the hot-forming approaches, such as a vacuum forming, vacuum pressure sky shaping, hot-platen pressure forming, and a deep-drawing vacuum forming.

[0008] After these containers insert contents, it carried out hot forming of a film, a sheet, or them for each other's opening, and a lid is pasted up, \*\* is used by being carried out in many cases, and thermal melting arrival is used also for this. Thus, a synthetic-resin film and a sheet use the property of thermal melting arrival, various processings are carried out, practical use is presented, and the adhesive strength and the appearance of the part which carried out thermal melting arrival on that occasion serve as an important property.

[0009] These containers, a film, and a sheet moreover, from the thermal resistance on storage and transportation Usually, the lightweight container which inserts the contents which the thermal resistance of 60 degrees C or more was needed, and were heated [ food / of structure length ], For example, in order that the hot philharmonic container used for the hood pack which packs food, such as rice and deep-fried dishes, in simple, a daily dish container or a jam, a pudding, and jelly may carry out \*\* after the thermal resistance in 80 degrees C or more is needed and it puts in contents, thermal melting arrival nature is needed.

[0010] In order to solve these heat-resistant problems, ebullition underwater and the approach of improving thermal resistance called \*\*\*\*\* in contraction from 66% to 4% by the heat test for 1 minute were shown to the U.S. Pat. No. 5,076,983 number official report by carrying out the heat set of the 130 degrees C of the oriented films of polylactic acid for 1 minute, but since it had already crystallized, this film had the trouble that it could not heat seal.

[0011]

[Problem(s) to be Solved by the Invention] The technical problem which this invention tends to solve is to provide various kinds of films, a sheet, a container, etc. with the lactic-acid system polymer layered product with the useful thermal resistance of 60 degrees C or more, the outstanding seal reinforcement, and biodegradability in which thermal melting arrival is possible.

[0012]

[Means for Solving the Problem] The result wholeheartedly examined in order that this invention persons might solve the above-mentioned technical problem, By using a lactic-acid system polymer layered product with the sealing layer (II) which consists of a lactic-acid system polymer (B) of the amorphism nature which has softening temperature lower than the melting point of the base material layer (I) which consists of a crystallized lactic-acid system polymer (A), and this lactic-acid system polymer (A), thermal melting arrival is possible. And it found out that a resin layered product with good thermal resistance could be offered, and this invention was completed.

[0013] Namely, this invention (1) The base material layer which consists of a crystallized lactic-acid system polymer (A) (I), The lactic-acid system polymer layered product which has the sealing layer (II) which consists of a lactic-acid system polymer (B) of the amorphism nature which has softening temperature lower than the melting point of a lactic-acid system polymer (A) and in which thermal melting arrival is possible, (2) A lactic-acid system polymer layered product given in (1) to which a lactic-acid system polymer (A) is characterized by being a lactic-acid system polymer with a melting point of 120 degrees C or more, [0014] (3) The polyester structural unit to which the lactic-acid system polymer (A) carried out dehydration condensation of dicarboxylic acid and the diol, And/or, a lactic-acid system polymer layered product given in (2) characterized by being the lactic-acid system polymer which includes the polyether structural unit which carried out dehydration condensation of dicarboxylic

acid and the polyether polyol three to 60% of the weight, (4) (2) to which a lactic-acid system polymer (A) is characterized by lactic-acid component content being 40% of the weight or more of a lactic-acid system polymer, or a lactic-acid system polymer layered product given in (3), (5) A lactic-acid system polymer layered product given in (2) to which a lactic-acid system polymer (A) is characterized by being polylactic acid, [0015] (6) A lactic-acid system polymer (A) and/or a lactic-acid system polymer (B) The lactic-acid system polymer layered product of any one publication of (1) - (5) characterized by consisting of the polymer to which deactivation of the polymerization catalyst was carried out by the quencher of a polymerization catalyst, (7) The lactic-acid system polymer layered product of any one publication of a lactic-acid system polymer (B) of softening temperature 40 - (1) - (6) characterized by being a 110-degree C lactic-acid system polymer, [0016] (8) The lactic-acid system polymer layered product of any one publication of (1) - (7) to which the base material layer (I) which consists of a lactic-acid system polymer (A) is characterized by crystallizing with a heat set, (9) The lactic-acid system polymer layered product of any one publication of (1) to the (8) to which the base material layer (I) which consists of a lactic-acid system polymer (A) is characterized by orientation being carried out by extension 1.4 to 16 times the field scale factor of this, (10) By the examining method (JIS-K-7198, A law) about the temperature dependence of dynamic viscoelasticity the melting point -- 20 -- degree C -- being low -- temperature -- the following -- a storage modulus (E') -- the minimum value -- 400 - 40,000 -- kg/cm<sup>2</sup> -- it is -- things -- the description -- \*\* -- carrying out -- (one) - (nine) -- some -- one -- a \*\* -- a publication -- a lactic acid -- a system -- a polymer -- a layered product -- [ -- 0017 -- ] (11) (1) - (the film using the lactic-acid system polymer layered product of any one publication of 10) or a sheet, and (12) (1) It is a container using the lactic-acid system polymer layered product of any one publication of - (10).

[0018]

[Embodiment of the Invention] This invention is a container which comes to carry out thermoforming of this layered product including the film which consists of the lactic-acid system polymer layered product which has the base material layer (I) which consists of a crystallized lactic-acid system polymer (A), and the sealing layer (II) which consists of a lactic-acid system polymer (B) of the amorphism nature which has softening temperature lower than the melting point of a lactic-acid system polymer (A), and in which thermal melting arrival is possible, and this layered product, a sheet a package bag, or a case

[0019] The base material layer (I) which consists of a lactic-acid system polymer (A) of this invention is a layer for realizing good thermal resistance, and consists of a crystallized lactic-acid system polymer. Moreover, the sealing layer (II) which consists of a lactic-acid system polymer (B) of amorphism nature is a layer for carrying out thermal melting arrival by the approach by heat sealing described previously, and consists of a lactic-acid system polymer which is the amorphism nature which has softening temperature lower than the melting point of the lactic-acid system polymer of a base material layer (I).

[0020] The lactic-acid system polymer of the amorphism nature said by this invention means what the peak of the melting point is not accepted in with the measuring method of JIS-K-7121. The lactic-acid system polymer (A) used for a base material layer (I) by this invention is the purpose which obtains good thermal resistance and thermoforming nature, and a lactic-acid system polymer with a melting point of 120-300 degrees C crystallized is used for it. As for the lactic-acid component as a constituent of a lactic-acid system polymer, as a lactic-acid system polymer suitable for this purpose, it is desirable that the ratio (ratio of length to diameter) of L bodies and D object which is the optical isomer of a lactic acid is the lactic-acid system polymer of 100 / 0 - 96/3 or 3 / 96 - 0/100.

[0021] A lactic-acid system polymer (A) includes the polyether structural unit which carried out dehydration condensation of the polyester structural unit and/or dicarboxylic acid which carried out dehydration condensation of dicarboxylic acid and the diol, and the polyether polyol three to 60% of the weight in the structural unit which carried out dehydration condensation of polylactic acid or the lactic acid. Moreover, as for weight average molecular weight, it is desirable that it is 10,000-500,000.

[0022] Moreover, since the lactic-acid system polymer (B) of the amorphism nature used for a sealing layer (II) realizes thermal melting arrival nature, 40-110 degrees C of softening temperatures of the softening temperature are a 40-100-degree C lactic-acid system polymer more preferably. Furthermore,

the ratio (ratio of length to diameter) of L bodies and D object of a lactic-acid component which are used is the lactic-acid system polymer of 95.5 / 4.5 - 4.5/95.5. A certain thing of the temperature gradient of the melting point of a lactic-acid system polymer (A) and the softening temperature of the lactic-acid system polymer (B) of amorphism nature crystallized here is [ from / after that the both-sexes ability of the thermal resistance of a lactic-acid system layered product and seal nature balances ] desirable 20 degrees C or more.

[0023] The polymers used for this invention are a lactic-acid system polymer containing the polyester which consists of polylactic acid and a lactic-acid component, a dicarboxylic acid component and a diol component, and/or a polyether polyol component, and its mixture, and lactic-acid component content is 40% of the weight or more of a thing. As a lactic-acid component, methylene chain length is mentioned for the thing of 2-10, for example, an adipic acid, a sebacic acid, a succinic acid, etc. as L and D which are each optical isomer, DL-lactic acid, and a dicarboxylic acid component.

[0024] As a diol component, the carbon number of a principal chain is mentioned for the thing of 2-6, for example, ethylene glycol, propylene glycol, a butylene glycol, 1 and 4-butanediol, 1, 6-hexandiol, etc. As polyether polyol, a polyethylene glycol, a polypropylene glycol, a polybutylene glycol, etc. are mentioned. It is most desirable to use a polypropylene glycol especially as propylene glycol and polyether polyol as an adipic acid, a sebacic acid, and a diol component as a dicarboxylic acid component also among these.

[0025] As a manufacturing method of polylactic acid, the lactide which is an annular dimer is compounded from a lactic acid, and although many methods of obtaining the polylactic acid of the amount of macromolecules by ring opening polymerization are used, the approach of compounding polylactic acid by direct dehydration condensation from a lactic acid is also used. Moreover, the copolymer which is a lactic-acid system polymer is obtained by adding the accessory constituent more than kinds, such as aliphatic series polyester, aromatic polyester, a caprolactone, vinyl acetate, an ethylene terephthalate polymer, and ethylene vinyl alcohol, and advancing a polymerization further immediately after the time of a polylactic acid polymerization, or a polylactic acid polymerization.

[0026] Moreover, although any phase of these polymerizations is sufficient, macromolecule quantification of the lactic-acid system polymer can be further carried out by adding macromolecule quantification agents, such as polyvalent carboxylic acid and/or its oxidation anhydride, and multiple-valued isocyanate. As polyvalent carboxylic acid, it is a succinic anhydride, trimellitic anhydride, pyromellitic dianhydride, etc. as trimellitic acid, pyromellitic acid, and an oxidation anhydride, and is the mixture of 2, 4-tolylene diisocyanate, 2, and 4-tolylene diisocyanate and 2 and 6-tolylene diisocyanate, and [0027] as multiple-valued isocyanate. There are diphenylmethane diisocyanate, 1,5-naphthalene diisocyanate, xylylene diisocyanate, hydrogenation xylylene diisocyanate, hexamethylene di-isocyanate, isophorone diisocyanate, a triphenylmethane color -4, 4', 4"-tri-isocyanate, etc. As an addition, the weight average molecular weight which usually goes up only to about 300,000 can be raised to 600,000 to about 700,000 about 0.01 to 5% of the weight to a lactic-acid system polymer that what is necessary is [ / just ].

[0028] Furthermore, the thermal stability of a polymer and storage stability can be raised by adding a chelating agent and alkyl acid phosphate as a quencher of a polymer polymerization catalyst after the time of this polymer polymerization, and/or polymerization termination. A chelating agent can suppress cutting of a polymer chain to min, and even if it mixes and uses an organic system chelating agent and an inorganic system chelating agent, it does not interfere.

[0029] The metal ion and complex in the catalyst included in the constituent which consists of polyester which serves as a polyhydroxy carboxylic acid from a dicarboxylic acid component and a diol component, and a macromolecule quantification agent as a device on the stable disposition by alkyl acid phosphate are formed, the activity is vanished, and the thermal stability of the constituent and storage stability are made to improve sharply.

[0030] As a chelating agent used, there are an organic system chelating agent and an inorganic system chelating agent. Especially as an organic system chelating agent, although not limited, N content carboxylic acid, a diazo compound, thiols, and porphyrins are mentioned as N content phenol and a

coordination atom as amino acid, phenols, hydroxycarboxylic acid, diketones, amines, an oxime, phenanthrolines, a pyridine compound, a dithio compound, and a coordination atom.

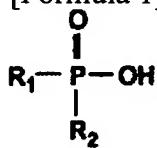
[0031] They form the metal ion and complex of a catalyst which are contained in a hydroxycarboxylic acid system polyester constituent, and make catalytic activity lose. Since effectiveness will be lost if hygroscopicity is high and absorbs moisture, an inorganic system chelating agent needs cautions for handling. Specifically, phosphoric acids, such as a phosphoric acid, phosphorous acid, a pyrophosphoric acid, and polyphosphoric acid, can be mentioned.

[0032] Although the addition of the organic system chelating agent of a chelating agent or an inorganic system chelating agent changes with the class of catalyst included in the class, a polyhydroxy carboxylic acid, and the polyester that consists of a dicarboxylic acid component and a diol component, and amounts, it is desirable to add the 0.1 - 100 weight section to 0.001 weight section - 5 weight section or the use catalyst 1 weight section to the total quantity of the polyester which serves as a polyhydroxy carboxylic acid from a dicarboxylic acid component and a diol component.

[0033] As alkyl acid phosphate; the mixture, such as alkyl acid phosphate, phosphonate, and alkyl phosphonic acid, is pointed out, and the general formula is shown in \*\* 1.

[0034]

[Formula 1]



[0035] (In R1, an alkyl group or an alkoxy group, and R2 express an alkyl group, an alkoxy group, or hydroxyl among a formula.)

The effectiveness to catalyst deactivation that alkyl acid phosphate is especially big is shown by alkyl acid phosphate. Although the addition of alkyl acid phosphate changes with the class, the class of use catalyst, and kneading conditions, it is desirable to add the 0.1 - 100 weight section to 0.001 weight sections - 5 weight section or the use catalyst 1 weight section to the total quantity of the polyester which serves as a polyhydroxy carboxylic acid from a dicarboxylic acid component and a diol component.

[0036] Especially the thing for which the lactic-acid system polymer which carried out deactivation of the polymerization catalyst by the quencher after the polymerization of a lactic-acid system polymer is used as a lactic-acid system polymer used for this invention is desirable. By deactivation processing of this polymerization catalyst, a residual monomer and oligomer decrease and thermal melting arrival nature improves.

[0037] The layered product as used in the field of this invention means a tabular thing with a thickness of 5000 micrometers or less. The lactic-acid system polymer layered product of this invention may also contain additives, such as other polymers, a plasticizer, a stabilizer, an anti-oxidant, an antiblocking agent, an antifogger, and a coloring agent, as the second and 3 component if needed. Aliphatic series polyester, polyvinyl alcohol, polyhydroxy butyrate-hydroxy BARIRETO, a starch system polymer, etc. may also be included in a polymer.

[0038] As an additive, plasticizers, such as polyester plasticizers, such as 1,3-butanediol and an adipic acid, and a dioctyl phthalate, a polyethylene-glycol adipic acid, epoxidized soybean oil, a stabilizer like a carbodiimide, 2, 6-Gee tertiary butyl-4-methyl phenol (BHT), an anti-oxidant like a butylhydroxyanisol (BHA), a silica, an antiblocking agent like talc, a glycerine fatty acid ester, an antifogger like citric-acid mono-stearyl, titanium oxide, carbon black, a coloring agent like ultramarine blue, etc. may be included.

[0039] Although there are the heat-sealing method, the impulse-heat-sealing method, the fusing seal method, the impulse fusing seal method, the ultrasonic seal method, the RF seal method, etc. in the adhesion approach using thermal melting arrival, generally the heat-sealing method, the impulse-heat-sealing method, and the fusing seal method are used well especially. Although the lactic-acid system

polymer layered product of this invention carries out the laminating of a base material layer (I) and the sealing layer (II), its co-extrusion forming-membranes method by two or more sets of extruders is [ the laminating approach ] the most practical. There is the approach of laminating the approach of carrying out the melting extrusion lamination of the sealing layer (II), the base material layer (I) which formed membranes beforehand, and a sealing layer (II) through adhesives in the base material layer (I) which otherwise formed membranes beforehand.

[0040] The metal or the metallic oxide may be vapor-deposited by the base material layer (I), printing may be performed, and two or more sorts of those processings may be carried out. Moreover, although the thickness of a base material layer (I) has desirable 5-3000 micrometers, 5-200 micrometers is more desirable from reinforcement and economical efficiency. Furthermore, it is desirable to make it the range of 5-100-micrometer thickness 100 micrometers or less called a film.

[0041] A sheet exceeds 100 micrometers and means the thing to 3000 micrometers here. Although 1-30 micrometers is desirable from the membrane formation nature of a film as thickness of a sealing layer (II), when a seal, and \*\* and economical efficiency are taken into consideration, 2-20 micrometers is 3-10 micrometers still more preferably more preferably.

[0042] The extrusion membrane formation approach of a lactic-acid system polymer and its condition are described. A lactic-acid system polymer has high hygroscopicity, since hydrolysis nature is also strong, it needs moisture management, and when carrying out extrusion molding using a common single screw extruder, it is necessary to absorb moisture [ polymer ] and dry this polymer by a vacuum dryer etc. before membrane formation. Moreover, since the dehydration effectiveness can skip a desiccation process highly, efficient membrane formation is possible for the membrane formation by the vent type biaxial extruder.

[0043] Although especially the melting extrusion temperature at the time of forming a lactic-acid system polymer is not restricted, the range of it is usually 150-250 degrees C. The sheet by which melting extrusion was carried out is cast so that it may become predetermined thickness, and it is cooled by the need. In that case, a touch roll, the Ayr knife, and when thin [ sheet thickness is thick and ], it considers as a uniform sheet by using electrostatic pinning properly. Although it is set to 0.2-3.0mm, if spacing of the lip which performs melting extrusion considers membrane formation nature, its 0.2-1.5mm will be desirable.

[0044] Next, although concrete explanation about the laminating approach is given, as the production approach of the lactic-acid system polymer layered product by co-extrusion membrane formation, melting and kneading of a base material layer (I) and a sealing layer (II) are first done with a separate extruder, a laminating is carried out within a T die or the feed block before it etc., and membranes are formed through a T die. The extrusion membrane formation approach and conditions follow the place described previously fundamentally.

[0045] When the adhesive property of a base material layer (I) and a sealing layer (II) is bad, it is necessary to set a glue line (III) to the interlayer. As resin used for this, the copolymer which introduced the special organic-functions machine, a butene system copolymer, polyethyleneimine, a denaturation cellulose, etc. are good for polyolefine etc. As thickness of a glue line (III), the range of 0.5-20 micrometers is desirable.

[0046] A melting extrusion lamination is the approach of pasting up a \*\*\*\*\* sealing layer (II) on the T die for laminators with a laminator, and laminating from the base material layer (I) and extruder which were sent with the delivery machine. The extrusion membrane formation approach and conditions of a sealing layer (II) follow the place described previously fundamentally. When the adhesive property of a base material layer (I) and a sealing layer (II) is bad, before sending a base material layer (I) to a laminator, adhesive improvement can be performed by surface irregularity processing of surface preparation, such as chemical etching processing of corona discharge treatment, frame plasma treatment, chromate treatment, etc., and ozone, ultraviolet treatment, sandblasting, etc. performing adhesive improvement, or choosing a suitable anchor coat agent.

[0047] A wet lamination, dry laminate, etc. are mentioned as the approach of the lamination which makes the base material layer (I) made beforehand and a sealing layer (II) rival. In this case, it is

necessary to laminate after applying adhesives in a base material layer (I) or a sealing layer (II). In a wet lamination, as adhesives, synthetic-resin molds, such as carbohydrate systems, such as protein systems, such as casein and gelatin, starch, and a cellulosic, vinyl acetate, acrylic ester, vinyl acetate of acrylic denaturation, and ethylene-vinyl acetate copolymerization resin, are mentioned.

[0048] The urethane system of the 2 liquid reaction type which mixes and uses base resin with the hydroxyl group of polyether system resin, such as polyester system resin, such as a 1 liquid reaction type which included the isocyanate radical in ends, such as polyether polyurethane poly isocyanate and polyester polyurethane poly isocyanate, as adhesives in the case of dry laminate, and polyester polyol, polyester polyurethane polyol, or polyether polyurethane polyol, and a curing agent with an isocyanate radical is mentioned. As coverage of these adhesives, about two 1 - 5 g/m is desirable.

[0049] The thermal resistance said by this invention means the practical thermal resistance which holds elasticity to some extent, and does not spoil an appearance or a film, a sheet, or its workpiece does not deform at the time of working temperature or use. If there is no temperature management also in the case of the usual transportation or storage in order to set workpieces, such as the bag and the case where a film, a sheet, or they were processed, and a lightweight container, in the sealing condition in a transport container and a warehouse etc., it is not rare to be exposed to ambient temperature 50 degrees C or more in summer. Therefore, the heat-resistant temperature of 60 degrees C or more is needed practically.

[0050] Although the lactic-acid system polymer (A) used for a lactic-acid system polymer base material layer (I) is crystallized in order to give thermal resistance, the heat setting method is explained as the heating art. When performing a heat set, you may carry out by the base material layer (I) independent which uses a lactic-acid system polymer (A), and may carry out in the state of the laminating of a base material layer (I) and a sealing layer (II).

[0051] Although not limited about temperature and especially time amount, it is desirable to make whenever [ stoving temperature ] into the range under of the melting point (Tm) from temperature lower 40 degrees C than the crystallization temperature (Tc) of a lactic-acid system polymer (A), for obtaining a proper crystallization rate. Especially, heat set temperature has especially the desirable range of temperature higher 40 degrees C than crystallization temperature (Tc) to it, in order to obtain a good field condition and good thermal resistance.

[0052] Furthermore, if extension processing is performed before a heat set or to coincidence, it can crystallization speed up and thermal resistance can be improved by the short heating processing time for about 5 - 30 seconds. Furthermore, since this is accompanied by crystallization by orientation, it can improve thermal resistance, with the good transparency of a lactic-acid system polymer held.

[0053] Although not restricted, immediately after especially this extension art carries out melting extrusion of the lactic-acid system polymer, rolling, vertical uniaxial stretching, horizontal uniaxial stretching, coincidence biaxial stretching, or serial biaxial stretching performs it on the sheet after preservation. Moreover, extension processing may be performed by the base material layer (I) independent which uses a lactic-acid system polymer (A), and may be performed in the state of the laminating of a base material layer (I) and a sealing layer (II).

[0054] As conditions, although the glass transition temperature (Tg) of a base material layer (I) to under the melting point is desirable and a temperature requirement high 50 degrees C is more desirable than especially glass transition temperature (Tg) from glass transition temperature, the field condition of a sheet is [ whenever / stoving temperature / in this case ] desirable [ a temperature requirement higher 10-40 degrees C especially than the glass transition temperature (Tg) of a base material layer (I) ] especially from a good thing. Although a field condition and transparency are good in the range it is 1.4 to 16 times whose field scale factor of this as draw magnification, the 2 to 16 times as many range as this is still more desirable.

[0055] The approach of making carry out fixed time amount contact on the approach of carrying out fixed time amount heating or a hot platen, metal mold, and a roll, and heating with radiant heat, such as air which carried out the forced convection, or an infrared heater, is mentioned to the heat set approach. The approach of using the equipment called especially a tenter can carry out the forced convection of the

heated air, can perform a heat set on a sheet or a film continuously, and is excellent in productivity. Since this equipment is equipment aiming at extension processing, it can do extension and a heat set for a short time, and is excellent in productivity. Crystallization of a lactic-acid system polymer layered product can be advanced efficiently.

[0056] Moreover, the heat set in the case of fabricating in the lightweight container which packs contents, such as food, a drink, a chemical, and miscellaneous goods, with rigidity for a lactic-acid system polymer layered product by the hot-forming approaches, such as a vacuum forming, vacuum pressure sky shaping, hot-platen pressure forming, and a deep-drawing vacuum forming, is good to carry out on the metal mold to fabricate. Although especially the die temperature and time amount that become a heat set condition at this time are not limited, they are chosen from the heat set temperature described previously.

[0057] It has the thermal resistance of 60 degrees C or more which whose lactic-acid system polymer layered product of this invention is in using at ordinary homes, and is satisfactory, and it is the examining method (JIS- K-7198, A law) about the temperature dependence of dynamic viscoelasticity, and the minimum value of the storage modulus (E') below temperature lower 20 degrees C than the melting point of a lactic-acid system polymer (A) is two or more 400 kgf/cm.

[0058] When the storage modulus (E') was smaller than 400 kgf/cm<sup>2</sup>, good elasticity cannot be acquired at 50-60 degrees C but contents are in a container, deformation is produced according to the load and contents are not supported. When the flexibility at the time of ordinary temperature use is also taken into consideration, as for a storage modulus (E'), it is desirable to adjust in the range of a maximum of 40,000 kgf/cm<sup>2</sup>. Furthermore, when acquiring the heat-resistant high temperature of 80 degrees C or more, as for a storage modulus (E'), it is more desirable to make it two or more 900 kgf/cm.

[0059] The trial about the temperature dependence of this dynamic viscoelasticity is performed by part for programming-rate/of 2 degrees C. The glass transition temperature (Tg) said by this invention, crystallization temperature (Tc), and the melting point (Tm) are Tig(s), Tpc(s), and Tpm(s) which are specified to JIS-K -7121, and perform a programming rate by part for 10-degree-C/. JIS-K-7121 are used for the lactic-acid system polymer of the amorphism nature said here, and it points out what the peak of the melting point is not accepted in. Measurement of softening temperature is performed by JIS-K -7206 and A law.

[0060] The lactic-acid system polymer layered product which has the sealing layer (II) which consists of a lactic-acid system polymer (B) of the amorphism nature which has softening temperature lower than the melting point of the base material layer (I) which consists of a lactic-acid system polymer (A) of this invention, and this lactic-acid system polymer (A) and in which thermal melting arrival is possible. Although a sealing surface, then good seal reinforcement are mutually obtained in a sealing layer (II), the effective seal reinforcement also as a sealing surface is obtained in a base material layer (I) and a sealing layer (II).

[0061] Moreover, the lactic-acid system polymer layered product in which the thermal melting arrival of this invention is possible can be used for the container with which general thermal resistance is demanded, and is suitable for the package bag aiming at a package or receipt, a case, and the manufacture of a lightweight container that comes to carry out thermoforming especially as a container.

[0062] It is the bag generally used, and a package bag is what carried out the seal of the synthetic-resin film by approaches, such as bending or adhesion, and is a three-dimensional packing-material gestalt here depending on superficial and the case. Although there are food, such as vegetables, confectionery, and a pan, miscellaneous goods or rice, fertilizer, etc. as an object of the package which used this, the lactic-acid system polymer layered product obtained here can be used for each [ these ] application as a package bag which bends or it comes to carry out thermal melting arrival.

[0063] a case -- bending processing of a sheet etc. -- box-like etc. -- or it is the three-dimensional packing material produced without processing it in three dimensions in the shape of a cylinder without bending and processing it, pasting up an edge by thermal melting arrival etc. depending on the case and being based on thermoforming, such as a vacuum forming and pressure forming. the object of the package which used this -- carrying out -- there are cosmetics, stationery, a small electrical-and-electric-

equipment product, a toy, miscellaneous goods, etc.

[0064] Moreover, there are some which contain superficially the document which opened the two way type which bends one side as another gestalt called case, carries out thermal melting arrival of other end faces, or carries out a two way type thermal melting arrival, and remains wide. The lactic-acid system polymer layered product obtained here can be used for each [ these ] application as a case which bends or it comes to carry out thermal melting arrival.

[0065] A lightweight container is a packing material fabricated in three dimensions using the thermoforming approaches, such as a vacuum forming, vacuum pressure sky shaping, hot-platen pressure forming, or a deep-drawing vacuum forming, in a film or a sheet. There is a cup filled up with a body, a lid, a tray or a hood pack, a blister pack, PTP, and a liquid according to the gestalt. As an object packed by the lightweight container, there are chemicals, such as drinks, such as food with which it is filled up, such as solid food, such as vegetables, meat, a daily dish, confectionery, a pan, and deep-fried dishes, or jelly, a jam, and a pudding, dairy products, and juice, and a tablet, miscellaneous goods, etc.

[0066] The lactic-acid system polymer layered product obtained by this invention has the outstanding thermal melting arrival nature and everyday thermal resistance, and can use them suitable for the packing material aiming at a package or receipt of various kinds of food, a drink, a chemical, miscellaneous goods, etc. especially a bag, a case, and the lightweight container that comes to carry out thermoforming.

[0067]

[Example] Although an example explains this invention to a detail, this invention is not limited to these. The production approach of the lactic-acid system polymer used for introduction this invention is shown.

[0068] (Production of a lactic-acid system polymer (A)) The tin octanoate 0.02 weight section was added for L-lactide 98 mol % and D-lactide 2 mol % after 1-hour churning and as an esterification catalyst under conditions with an inside [ of an inert gas ambient atmosphere ], and a temperature of 165 degrees C, and the reaction was performed for 8 hours. Then, the alkyl-acid-phosphate 0.04 weight section was added and kneaded as a quencher. The obtained lactic-acid system polymer (A) was transparent and colorless resin, and, for weight average molecular weight, 192,000 and glass transition temperature (Tg) were [ 110 degrees C and the melting point (Tm) of 56 degrees C and crystallization temperature (Tc) ] 163 degrees C from the measurement result of GPC. This resin is called P1.

[0069] The 0.02 weight section, in addition a 8-hour reaction were performed [ L-lactide ] for tin octanoate as an esterification catalyst after 1-hour churning under conditions with a temperature of 185 degrees C among the inert gas ambient atmosphere. Then, the alkyl-acid-phosphate 0.04 weight section was added and kneaded as a quencher. The obtained lactic-acid system polymer (A) was transparent and colorless resin, and, for weight average molecular weight, 252,000 and glass transition temperature (Tg) were [ 110 degrees C and the melting point (Tm) of 59 degrees C and crystallization temperature (Tc) ] 176 degrees C from the measurement result of GPC. This resin is called P2.

[0070] the aliphatic series system polyester (weight average molecular weight: 24,000 (polystyrene conversion) and 50 mol % [ of sebacic acids ], propylene glycol 50 mol %) 5 weight section -- the lactide (L-lactide 99 mol %, D-lactide 1 mol %) 95 weight section -- in addition, inert gas permuted the ambient atmosphere, it was made to mix at 170 degrees C for 1 hour, and the 0.02 weight section, in addition a 8-hour reaction were performed for tin octanoate as an esterification catalyst. Then, the alkyl-acid-phosphate 0.04 weight section was added and kneaded as a quencher. The obtained lactic-acid system polymer (A) was transparent and colorless resin, and, for weight average molecular weight, 203,000 and glass transition temperature (Tg) were [ 108 degrees C and the melting point (Tm) of 55 degrees C and crystallization temperature (Tc) ] 168 degrees C from the measurement result of GPC. This resin is called P3.

[0071] the aliphatic series system polyester (weight average molecular weight: 35,000 (polystyrene conversion) and 50 mol % [ of sebacic acids ], 1,6-hexanediol 50 mol %) 20 weight section -- the lactide (L-lactide 97 mol %, D-lactide 3 mol %) 80 weight section -- in addition, inert gas permuted the ambient atmosphere, it was made to mix at 170 degrees C for 1 hour, the tin octanoate 0.02 weight section was

added as an esterification catalyst, and the reaction was performed for 8 hours. Then, the alkyl-acid-phosphate 0.04 weight section was added and kneaded as a quencher. the obtained lactic-acid system polymer (A) was transparent and colorless resin, weight average molecular weight came out of 113,000 and glass transition temperature (Tg) from the measurement result of GPC, and 49 degrees C and crystallization temperature (Tc) came out of 93 degrees C and 162 degrees C (Tm) of melting points. This resin is called P4.

[0072] the aliphatic series system polyester (weight average molecular weight: 34,000 (polystyrene conversion) and 50 mol % [ of sebacic acids ], ethylene glycol 25 mol %, 1,6-hexanediol 25 mol %) 40 weight section -- the lactide (L-lactide 97 mol %, D-lactide 3 mol %) 60 weight section -- in addition, inert gas permuted the ambient atmosphere, it was made to mix at 165 degrees C for 1 hour, the tin octanoate 0.02 weight section was added as an esterification catalyst, and the reaction was performed for 8 hours. Then, the alkyl-acid-phosphate 0.04 weight section was added and kneaded as a quencher. the obtained lactic-acid system polymer (A) was transparent and colorless resin, weight average molecular weight came out of 102,000 and glass transition temperature (Tg) from the result of GPC, and 51 degrees C and crystallization temperature (Tc) came out of 73 degrees C and 162 degrees C (Tm) of melting points. This resin is called P5.

[0073] (Production of the lactic-acid system polymer (B) of amorphism nature) The 0.02 weight section, in addition a 8-hour reaction were performed [ L-lactide 70 mol % and D-lactide 30 mol % ] for tin octanoate as an esterification catalyst after 1-hour churning under conditions with a temperature of 165 degrees C among the inert gas ambient atmosphere. Then, the alkyl-acid-phosphate 0.04 weight section was added and kneaded as a quencher. The obtained lactic-acid system polymer (B) was transparent and colorless resin, 272,000 was not seen for weight average molecular weight from the measurement result of GPC, and 52 degrees C and the melting point (Tm) were not seen for glass transition temperature (Tg). This resin is called P6.

[0074] The 0.02 weight section, in addition a 8-hour reaction were performed [ L-lactide 30 mol % and D-lactide 70 mol % ] for tin octanoate as an esterification catalyst after 1-hour churning under conditions with a temperature of 165 degrees C among the inert gas ambient atmosphere. Then, the alkyl-acid-phosphate 0.04 weight section was added and kneaded as a quencher. The obtained lactic-acid system polymer (B) was transparent and colorless resin, 167,000 was not seen for weight average molecular weight from the measurement result of GPC, and 52 degrees C and the melting point (Tm) were not seen for glass transition temperature (Tg). This resin is called P7.

[0075] the aliphatic series system polyester (weight average molecular weight: 75,000 (polystyrene conversion) and 50 mol % [ of sebacic acids ], ethylene glycol 50 mol %) 50 weight section -- the lactide (L-lactide 90 mol %, D-lactide 10 mol %) 50 weight section -- in addition, inert gas permuted the ambient atmosphere, it was made to mix at 165 degrees C for 1 hour, and the 0.02 weight section, in addition a 8-hour reaction were performed for tin octanoate as an esterification catalyst. Then, the alkyl-acid-phosphate 0.04 weight section was added and kneaded as a quencher. The obtained lactic-acid system polymer (B) was transparent and colorless resin, 81,000 was not seen for weight average molecular weight from the measurement result of GPC, and 46 degrees C and the melting point (Tm) were not seen for glass transition temperature (Tg). This resin is called P8.

[0076] Polyester containing aromatic carboxylic acid and aliphatic series dicarboxylic acid (weight average molecular weight 47,000 (polystyrene conversion)) 16 mol % and 14 mol [ of isophthalic acid ] % and 20 mol [ of adipic acids ] % of terephthalic acids, ethylene glycol 23 mol % and the neopentyl glycol 27 mol % 5 weight section -- lactide (L-lactide 95 mol % --) The D-lactide 5 mol % 95 weight section was added, inert gas permuted the ambient atmosphere, melting and mixing of both were done at 165 degrees C for 1 hour, and the reaction was performed for tin octanoate as an esterification catalyst for 0.02 \*\*\*\*\* 6 hours. Then, the alkyl-acid-phosphate 0.04 weight section was added and kneaded as a quencher. The obtained lactic-acid system polymer (B) is transparent and colorless resin, and 57 degrees C and the melting point (Tm) were not seen for weight-average-molecular-weight 195,000 and glass transition temperature (Tg). This resin is called P9.

[0077] The 0.02 weight section, in addition a 8-hour reaction were performed [ L-lactide 70 mol % and

D-lactide 30 mol % ] for tin octanoate as an esterification catalyst after 1-hour churning under conditions with a temperature of 165 degrees C among the inert gas ambient atmosphere. Then, the quencher did not add. The obtained lactic-acid system polymer (B) was transparent and colorless resin, 272,000 was not seen for weight average molecular weight from the measurement result of GPC, and 52 degrees C and the melting point (Tm) were not seen for glass transition temperature (Tg). This resin is called P10.

[0078] Polyester containing aromatic carboxylic acid and aliphatic series dicarboxylic acid (weight average molecular weight 47,000 (polystyrene conversion)) 16 mol % and 14 mol [ of isophthalic acid ] % and 20 mol [ of adipic acids ] % of terephthalic acids, ethylene glycol 23 mol % and the neopentyl glycol 27 mol %5 weight section -- lactide (L-lactide 95 mol % --) The D-lactide 5 mol %95 weight section was added, inert gas permuted the ambient atmosphere, melting and mixing of both were done at 165 degrees C for 1 hour, and the reaction was performed for tin octanoate as an esterification catalyst for 0.02 \*\*\*\*\* 6 hours. The quencher did not add. The obtained lactic-acid system polymer (B) is transparent and colorless resin, and 57 degrees C and the melting point (Tm) were not seen for weight-average-molecular-weight 195,000 and glass transition temperature (Tg). This resin is called P11.

[0079] (Examples 1-5) The lactic-acid system polymer (A) shown in Table 1 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of a base material layer (I), (20 micrometers), and the laminated film with a thickness [ of a sealing layer (II) (15 micrometers) configuration ] of 35 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). The heat set for 10 minutes was performed on the film in 100-degree C air oven after that. The next evaluation was carried out in the obtained laminated film.

[0080] (1) the sealing layer of the laminated film of two measurement profit \*\*\*\* of seal reinforcement was doubled mutually, it considered as the sealing surface, and thermal melting arrival was carried out by the heat sealer (product made from circuit tester industry). Then, tension test 180-degree friction test was carried out with the machine (Shimadzu Corp. make), and the bond strength of the thermal melting arrival by the heat-sealing method was measured as seal reinforcement. A result is shown in Table 1.

[0081] Seal conditions: The seal bar temperature of 60-140 degrees C, seal \*\* 2 kgf/cm<sup>2</sup>, seal time amount 1 second (the used silver dimension is 10x300mm)

Seal measurement on the strength: It is based on JIS-Z -1707.

[0082] (2) Heat-resistant evaluation \*\* real trial : the obtained laminated film was cut down in the magnitude of 20cm around, 300g weight was placed in the center, four corners were together banded so that it might be wrapped, and the simple trial bag was produced. Suspension of this was carried out so that a union part might turn up into the air oven (60 degrees C and 80 degrees C) of each test temperature, and the condition of the film under the effect of the weight of 20 minutes after was observed. The die length of the trial bag at the time of suspension is 13.5cm. What especially change was not regarded as is O (the die length of a trial bag is a less than 14cm thing). The thing (that to which the die length of a trial bag exceeds 15cm) which developed remarkably with weight and deformed, or the thing to which bag tearing was carried out and weight fell is x. The condition in the meantime was evaluated as \*\*. A result is shown in Table 1.

[0083] \*\* Measurement of a storage modulus (E') : the minimum value was calculated using JIS-K-7198 and A law from measurement of the storage modulus (E') below temperature lower 20 degrees C than the melting point of a crystalline lactic-acid system polymer (A). A result is shown in Table 1.

[0084] (3) Hayes value : JIS-K -7105 and a result are shown in Table 1.

(4) The melting point of resin : JIS-K -7121 and a result are shown in Table 1.

(5) Softening temperature of resin : JIS-K -7206 and a result are shown in Table 1.

[0085] (6) Biodegradability evaluation : 5kg of kitchen garbages was put into outdoor compost (capacity of 100l.), and the test piece of 10cm around started from the laminated film obtained on it was placed. Viewing estimated the condition of the test piece one month after carrying a kitchen garbage with a thickness of about 5 morecm. In addition, this trial was performed in the summer. A valuation basis is as follows. There is degradation of physical properties remarkably and it is O which has difficult

maintenance of a configuration. It is \*\* which is maintaining the configuration although there are deformation and milkiness. What there are not milkiness, deformation, etc. and is maintaining the condition before test initiation was taken as x.

[0086] Seal initiation temperature is near 80 degree C, and each laminated film has 1000gf(s) / good seal reinforcement of 15mm or more in 90 degrees C or more. About thermal resistance, it was good in the real trial which used weight at the test temperature of 60 degrees C and 80 degrees C. At this time, the minimum value of the storage modulus (E') of each laminated film is two or more 900 kgf/cm. Moreover, the produced lactic-acid system polymer layered product was transparency, and biodegradability was good.

[0087] (Examples 6 and 7) The lactic-acid system polymer (A) shown in Table 1 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of a base material layer (I), (20 micrometers), and the laminated film with a thickness [ of a sealing layer (II) (15 micrometers) configuration ] of 35 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). The heat set for 10 minutes was performed on the film in 100-degree C air oven after that. The next evaluation was carried out in the obtained laminated film.

[0088] (1) Measurement of seal reinforcement : the sealing layer and base material layer of the laminated film of two sheets which were obtained were doubled mutually, it considered as the sealing surface, and thermal melting arrival was carried out by the heat sealer (circuit tester industrial company make). Then, 180-degree friction test was carried out with the tension tester (Shimadzu Corp. make), and the bond strength of the thermal melting arrival by the heat-sealing method was measured as seal reinforcement. A result is shown in Table 1.

[0089] Seal conditions: The seal bar temperature of 60-140 degrees C, seal \*\* 2 kgf/cm<sup>2</sup>, seal time amount 1 second (the used silver dimension is 10x300mm)

seal on the strength measurement: -- JIS-Z-1707(2) thermal-resistance evaluation: -- the approach performed in the examples 1-5 -- the same . A result is shown in Table 1.

(3) Hayes value : JIS-K-7105 and a result are shown in Table 1.

(4) The melting point of resin : JIS-K-7121 and a result are shown in Table 1.

(5) Softening temperature of resin : JIS-K-7206 and a result are shown in Table 1.

[0090] (6) biodegradability trial: -- the approach performed in the examples 1-5 -- the same . A result is shown in Table 1.

Each laminated film has the seal reinforcement around 160 - 180gf / 15mm in 90 degrees C or more. About thermal resistance, it was satisfactory in the real trial which used weight at the test temperature of 60 degrees C and 80 degrees C. At this time, the minimum value of the storage modulus (E') of each laminated film is two or more 900 kgf/cm. The produced lactic-acid system layered product had transparency and good biodegradability.

[0091] (Examples 8 and 9) The lactic-acid system polymer (A) shown in Tables 1 and 2 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of a base material layer (I), (80 micrometers), and the laminating sheet with a thickness [ of a sealing layer (II) (60 micrometers) configuration ] of 140 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). next, the 35-micrometer oriented film was produced by the single-engined biaxial drawing machine (the Iwamoto factory company make) for the extension temperature of 65 degrees C, and preheating-time 5 minutes on condition that the part [ for /], and 100% draw magnification 2of extension rates x2(vertical x width): side scale factor 4. On both sides of the film, it fixed to the frame after that, and the heat set for 20 seconds was performed in 100-degree C air oven.

[0092] While measuring the seal reinforcement of the obtained laminated film, thermal resistance, Hayes, and biodegradability were performed like the approach which evaluated by examples 1-5. The result is shown in Tables 1 and 2. Seal initiation temperature is near 80 degree C, and each laminated film has 1000gf(s) / good seal reinforcement of 15mm or more in 90 degrees C or more. About thermal resistance, it was satisfactory in the example 8 in the real trial which used weight at the test temperature

of 60 degrees C and 80 degrees C. In the example 9, the elongation of some film was seen with the test temperature of 80 degrees C. At this time, the minimum value of the storage modulus (E') of each laminated film is 900 kgf/cm<sup>2</sup> in the example 8, and is 600 kgf/cm<sup>2</sup> in the example 9. The transparency of the film produced here and biodegradability were good.

[0093] (Example 10) The lactic-acid system polymer (A) shown in Table 2 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of two sorts of each monolayer films with a thickness of 25 micrometers was carried out using the extruder (the Tanabe plastics company make). The heat set only of the monolayer film which consists of a lactic-acid system polymer (A) here was carried out for 10 minutes in 100-degree C air oven. Then, desiccation was performed, after applying the gelatin solution to one side of these two sorts of films 30%, respectively and being stuck by pressure by 2 kgf/cm<sup>2</sup>. Consequently, the good laminated film of an appearance was obtained. It was similarly estimated as the approach which performed the seal reinforcement of the obtained laminated film, thermal resistance, Hayes, and biodegradability in the examples 1-5. The result is shown in Table 2.

[0094] Seal initiation temperature is near 80 degree C, and the produced laminated film has 1000gf(s) / good seal reinforcement of 15mm or more in 90 degrees C or more. About thermal resistance, it was satisfactory in the real trial which used weight at the test temperature of 60 degrees C and 80 degrees C. The minimum value of the storage modulus (E') of a laminated film is two or more 900 kgf/cm 1100 Kgf/cm<sup>2</sup>. The film produced here had transparency and good biodegradability.

[0095] (Example 11) The lactic-acid system polymer (A) shown in Table 2 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), a sealing layer (II) and (15 micrometers) were formed in base material layer (I) (20 micrometers) both sides using the co-extrusion machine (the Tanabe plastics company make), and extrusion membrane formation of the 50-micrometer laminated film was carried out. then, this film -- the inside of 100-degree C air oven -- the heat set for 10 minutes -- it carried out. It was similarly estimated as the approach which performed the seal reinforcement of the obtained laminated film, thermal resistance, Hayes, and biodegradability in the examples 1-5. A result is shown in Table 2.

[0096] The seal initiation temperature of the film concerned is near 80 degree C, and has 1000gf(s) / good seal reinforcement of 15mm or more in 90 degrees C or more. Although it was satisfactory in the real trial which used weight about thermal resistance at the test temperature of 60 degrees C, the elongation of a film was seen at 80 degrees C. The minimum value of the storage modulus (E') of each laminated film is 780 kgf/cm<sup>2</sup>. The transparency of a film and biodegradability were good.

[0097] (Example 12) The lactic-acid system polymer (A) shown in Table 2 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of a base material layer (I), (20 micrometers), and the laminated film with a thickness [ of a sealing layer (II) (15 micrometers) configuration ] of 35 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). The heat set was performed for the film for 10 minutes in 100-degree C air oven after that. The end of the laminated film obtained considering the sealing layer as an inside of a package bag here was turned up, and the bottom part of a package bag was formed. Next, the package bag of 20cm around which heat sealed the both sides of a part using the heat sealer (product made from circuit tester industry) by return, and the method of one opened wide was produced. The appearance of the produced package bag is good. The seal reinforcement of the methods of two which carried out the seal was also good. Seal conditions: The seal bar temperature of 90 degrees C, seal \*\* 2 kgf/cm<sup>2</sup>, seal time amount 1 second (the used silver dimension is 10x300mm)

[0098] Heat-resistant evaluation put 300g weight into the produced package bag, was made into suspension into the air oven (60 degrees C and 80 degrees C) of each test temperature, and was performed by observing the condition of the package bag under the effect of the weight of 20 minutes after. The die length of the package bag at the time of suspension is about 20cm. What especially change was not regarded as is O (the die length in a bag is a less than 21cm thing). The thing which developed remarkably with weight and deformed, or the thing to which bag tearing was carried out and weight fell

is x (that to which the die length in a bag exceeds 22cm). The condition in the meantime was evaluated as \*\*. Consequently, especially deformation and a tear were not looked at by the package bag.

[0099] Hayes value: It was similarly estimated as the approach performed in the examples 1-5 about the obtained film. A result is shown in Table 2. Biodegradability evaluation was similarly estimated as the approach performed in the examples 1-5 by using as a test piece the package bag in which the kitchen garbage was put. Biodegradability was good.

[0100] (Example 13) The lactic-acid system polymer (A) shown in Table 2 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of a base material layer (I), (20 micrometers), and the laminated film with a thickness [ of a sealing layer (II) (15 micrometers) configuration ] of 35 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). The heat set of the film was carried out for 10 minutes in 100-degree C air oven after that. The end of the laminated film obtained considering the sealing layer as an inside of a package bag here was turned up, and the bottom part of a package bag was formed. Next, the package bag of 20cm around which carried out the fusing seal of the both sides of a part by return, and the method of one opened wide was produced. The appearance of the produced package bag is good. The seal reinforcement of the methods of two which carried out the seal was also good.

seal condition: -- seal bar temperature Hayes value [ of 300 degrees C ]: -- it was similarly estimated as the approach performed in the examples 1-5 about the obtained film. A result is shown in Table 2.

[0101] Heat-resistant evaluation was performed by the same approach as an example 12. Consequently, it is [ especially no deformation and tear ] in a package bag and was good at each test temperature of 60 degrees C and 80 degrees C. Biodegradability evaluation was performed by the same approach as an example 12. Consequently, biodegradability was good.

[0102] (Example 14) the lactic-acid system polymer (A) shown in Table 2 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of the laminating sheet with a thickness [ of a base material (layer I) (20 micrometer) sealing layer (II):(15 micrometers) configuration ] of 35 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). The heat set was carried out to the sheet for 10 minutes in 100-degree C air oven after that. The end of the laminated film obtained considering the sealing layer as an inside of a package bag here was turned up, and the bottom part of a package bag was formed. Next, the package bag of 20cm around which carried out impulse heat sealing of the both sides of a part by return, and the method of one opened wide was produced.

[0103] The appearance of the produced package bag is good. The seal reinforcement of the methods of two which carried out the seal was also good. The sealing machine used the Fuji poly sealer. About thermal resistance, it examined by the same approach as an example 12. Consequently, especially deformation and a tear were not looked at by the package bag with each test temperature of 60 degrees C and 80 degrees C. Biodegradability evaluation was performed by the same approach as an example 12. Consequently, biodegradability was good.

[0104] (Example 15) The lactic-acid system polymer (A) shown in Table 2 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of a base material layer (I), (100 micrometers), and the laminating sheet with a thickness [ of a sealing layer (II) (15 micrometers) configuration ] of 115 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). an after that film -- the inside of 100-degree C air oven -- the heat set for 10 minutes -- it carried out. The laminating sheet obtained considering the sealing layer as an inside of a case here was rounded off so that it might become a cylinder, superposition heat sealing of the both ends was carried out, and the side-face part of a cylinder-like case was produced. Cylindrical height is 10cm and a diameter is 5cm. The appearance of the seal part of the produced case side face is good. Seal reinforcement was also good.

Seal conditions: The seal bar temperature of 90 degrees C, seal \*\* 2 kgf/cm<sup>2</sup>, seal time amount 1 second.

[0105] The cylindrical case produced [ which produced and thermal-resistance-evaluated ] was stood perpendicularly in the air oven (60 degrees C and 80 degrees C) of each test temperature, was set for 20 minutes, and was performed. Contraction, deformation, etc. were not especially seen. Biodegradability evaluation was similarly estimated as the approach performed in the examples 1-5 by using as a test piece the case in which the kitchen garbage was put. Consequently, biodegradability was good.

[0106] (Example 16) The lactic-acid system polymer (A) shown in Table 2 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of a base material layer (I), (250 micrometers), and the laminating sheet for vacuum formings with a thickness [ of a sealing layer (II) (15 micrometers) configuration ] of 265 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). Moreover, extrusion membrane formation of the laminated film for seals with a thickness [ of base material (layer I):20micrometer and a sealing layer (II):15 micrometer configuration ] of 35 micrometers was carried out similarly. The film carried out the heat set for 10 minutes in 100-degree C air oven after that.

[0107] The contraction ratio performed the vacuum forming with the cup metal mold of 0.36 (female mold) by the round shape whose diameter of opening is 55mm, and the sheet fabricated it in the lightweight container for evaluation. The sealing layer was made into the container inside at this time, and width of face of the flange of the mold goods equivalent to a seal part was set to 3mm.

Vacuum-forming conditions: Heater temperature of 400 degrees C. Heating time 10 seconds. The die temperature of 80 degrees C. Cycle time 30 seconds.

[0108] Heat-resistant evaluation was performed by setting for 20 minutes in the produced cup in the air oven (60 degrees C and 80 degrees C) of each test temperature. Especially in 60 degrees C, although contraction, deformation, etc. were not seen, some deformation was seen at 80 degrees C. Evaluation is appearance evaluation (O: fitness, x:contraction, deformation of a \*\*:some). Next, the cup and the film for seals were heat sealed using the auto cup sealer (the Sunrise system company make) in the seal temperature of 90 degrees C, seal \*\* 2 kgf/cm<sup>2</sup>, and seal time amount 1 second. The sealing surface was taken as the mutual sealing layer. The seal reinforcement of the lightweight container which carried out the seal was good, and its appearance which carried out the seal was also good.

[0109] Biodegradability evaluation was similarly estimated as the approach which performed the test piece in the examples 1-5 as a lightweight container in which the kitchen garbage was put. Consequently, biodegradability was good.

[0110] (Examples 17 and 18) The lactic-acid system polymer (A) shown in Table 3 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of a base material layer (I), (20 micrometers), and the laminated film with a thickness [ of a sealing layer (II) (15 micrometers) configuration ] of 35 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). The heat set for 10 minutes was performed on the film in 100-degree C air oven after that. The next evaluation was carried out in the obtained laminated film. It was similarly estimated as the approach which performed the seal reinforcement of the obtained laminated film, thermal resistance, Hayes, and biodegradability in the examples 1-5. The result is shown in Table 3.

[0111] The seal initiation temperature of the film concerned is near 80 degree C, and shows 500gf / good seal reinforcement of 15mm or more in 90 degrees C or more. About thermal resistance, it was good in the real trial which used weight at the test temperature of 60 degrees C and 80 degrees C. At this time, the minimum value of the storage modulus (E') of each laminated film was 1000 kgf/cm<sup>2</sup> in 1300 kgf/cm<sup>2</sup> and an example 18 in the example 17. The transparency of a film and biodegradability were good. In addition, seal temperature was indicated beside the seal reinforcement in Table 1-4.

[0112]

[Table 1]

実施例		1	2	3	4	5	6	7	8
乳酸系ポリマー (A)	P1	P2	P3	P4	P5	P3	P4	P1	
融点 [℃]	163	176	168	162	162	163	162	163	
結晶化温度 [℃]	110	110	108	93	73	108	93	110	
乳酸系ポリマー (B)	P6	P7	P8	P9	P9	P8	P9	P8	
軟化温度 [℃]	52	52	46	57	57	46	57	46	
厚み [μm]	35	35	35	35	35	35	35	35	
ヘイズ [%]	13	18	10	19	14	10	19	1.0	
耐 熱 性	60℃	○	○	○	○	○	○	○	○
	80℃	○	○	○	○	○	○	○	○
E' [kgf/cm²]	1300	1500	1300	1100	1000	1300	1100	900	
生分解性	○	○	○	○	○	○	○	○	
シール強度 [gf/15mm]	80℃	80	70	30	500	500	10	10	0
	90℃	1400	1350	1400	1450	1350	170	160	300
	100℃	1700	1750	1600	1600	1650	180	170	1400
	120℃	2000	1900	1900	1650	1650	180	170	1550
	140℃	2000	1900	2000	1650	1700	180	170	1700

[0113]  
[Table 2]

実施例		9	10	11	12	13	14	15	16
乳酸系ポリマー (A)	P5	P4	P1	P2	P3	P4	P2	P3	
融点 [℃]	162	162	163	176	168	162	176	168	
結晶化温度 [℃]	73	93	110	110	108	93	110	108	
乳酸系ポリマー (B)	P9	P9	P6	P7	P8	P9	P7	P8	
軟化温度 [℃]	57	57	52	52	46	57	52	46	
厚み [μm] ヘイズ [%]	35 1.1	50 15	35 18	35 18	35 10	35 21	— —	— —	
耐 熱 性	実試験 60°C	○	○	○	○	○	○	○	○
	80°C	△	○	△	○	○	○	○	△
E' [kgf/cm²]		600	1100	780	1500	1300	1100	1500	—
生分解性		○	○	○	○	○	○	○	○
シール強度 [gf/15mm]	80°C	0	400	80					
	90°C	600	1350	1400					
	100°C	1650	1900	1700					
	120°C	1900	2300	2000					
	140°C	1900	2350	2000					

[0114]  
[Table 3]

実施例		17	18
乳酸系ポリマー (A)		P1	P4
融点 [℃]		163	162
結晶化温度 [℃]		110	93
乳酸系ポリマー (B)		P10	P11
軟化温度 [℃]		52	57
厚み [μm]		35	35
ヘイズ [%]		13	19
耐熱性 実試験	60℃	○	○
	80℃	○	○
	E' [kgf/cm²]	1300	1100
生分解性		○	○
シール強度 [gf/15mm]	80℃	40	200
	90℃	600	700
	100℃	800	800
	120℃	1000	800
	140℃	1000	800

[0115] The lactic-acid system polymer (B) of amorphism nature shown in Table 4 (Examples 1 and 2 of a comparison) In a base material layer (I) A lactic-acid system polymer (A) is made into a sealing layer (II), and a co-extrusion machine (the Tanabe plastics company make) is used. A base material layer (I), (20 micrometers), Extrusion membrane formation of the laminated film with a thickness [ of a sealing layer (II) (15 micrometers) configuration ] of 35 micrometers was carried out, and the heat set of the membrane formation film was carried out for 10 minutes in 100-degree C air oven. It was similarly estimated as the approach which performed the seal reinforcement of the obtained laminated film, thermal resistance, Hayes, and biodegradability in the examples 1-5. The result is shown in Table 4. In the real trial for which each laminated film used weight about thermal resistance, it was satisfactory at the test temperature of 60 degrees C and 80 degrees C. At this time, the minimum value of the storage modulus (E') of a laminated film was two or more 900 kgf/cm. Seal reinforcement was not obtained.

[0116] (Examples 3 and 4 of a comparison) The lactic-acid system polymer (B) of amorphism nature shown in Table 4 was made as the base material layer, the lactic-acid system polymer (A) was made into the sealing layer (II), and extrusion membrane formation of a base material layer (I), (80 micrometers), and the laminating sheet with a thickness [ of a sealing layer (II) (60 micrometers) configuration ] of 140 micrometers was carried out. next, the 35-micrometer extension laminated film was produced by the single-engined biaxial drawing machine (the Iwamoto factory company make) for the extension temperature of 65 degrees C, and preheating-time 5 minutes on condition that the part [ for /], and 100% draw magnification 2of extension rates x2(vertical x width): side scale factor 4. It fixed to the frame of 30cm angle on both sides of the film, and the heat set was carried out for 20 seconds in 100-degree C air

oven. It was similarly estimated as the approach which performed the seal reinforcement of the obtained laminated film, thermal resistance, Hayes, and biodegradability in the examples 1-5. The result is shown in Table 4.

[0117] Although it was satisfactory at the test temperature of 60 degrees C in the real trial for which each laminated film used weight about thermal resistance, the elongation of a film was seen at 80 degrees C. At this time, the minimum value of the storage modulus (E') of each laminated film was 500 kgf/cm<sup>2</sup> in 600 kgf/cm<sup>2</sup> and the example 4 of a comparison in the example 3 of a comparison. However, seal reinforcement was not obtained.

[0118] (Example 5 of a comparison) The lactic-acid system polymer (B) of amorphism nature shown in Table 4 was made as the base material layer (I), the lactic-acid system polymer (A) was made into the sealing layer (II), and extrusion membrane formation of two sorts of each monolayer films with a thickness of 25 micrometers was carried out using the extruder (the Tanabe plastics company make). The monolayer film made from the lactic-acid system polymer (B) here performed the heat set for 10 minutes in 100-degree C air oven. It was made to dry, after applying gelatin to one side of the obtained film of two sheets 30% and being stuck by pressure by 2 kgf/cm<sup>2</sup>.

[0119] Consequently, the laminated film with a good thickness [ of an appearance ] of about 50 micrometers was obtained. It was similarly estimated as the approach which performed the seal reinforcement of the obtained laminated film, thermal resistance, Hayes, and biodegradability in the examples 1-5. The result is shown in Table 4. About thermal resistance, deformation by remarkable elongation was seen with the test temperature of 60 degrees C and 80 degrees C in the real trial which used weight. Seal reinforcement was not obtained although the minimum value of the storage modulus (E') of a laminated film was two or more 900 kgf/cm 1100 kgf/cm<sup>2</sup>.

[0120] (Example 6 of a comparison) The lactic-acid system polymer (A) shown in Table 4 was made as the base material layer (I), the lactic-acid system polymer (B) of amorphism nature was made into the sealing layer (II), and extrusion membrane formation of the laminated film with a thickness [ of base material (layer I):20micrometer and a sealing layer (II):15 micrometer configuration ] of 35 micrometers was carried out using the co-extrusion machine (the Tanabe plastics company make). Then, it was similarly estimated as the approach which performed the thermal resistance of the laminated film obtained without performing a heat set, and Hayes in the examples 1-5. A result is shown in Table 4. About the thermal resistance of the produced laminated film, the tear was seen with the test temperature of 60 degrees C and 80 degrees C in the real trial which used weight. The minimum value of the storage modulus (E') of a laminated film is 0 kgf/cm<sup>2</sup>, and there was no thermal resistance.

[0121] (Example 7 of a comparison) After using the lactic-acid system polymer (A) shown in Table 4 and carrying out extrusion membrane formation of the monolayer film with a thickness of 35 micrometers using an extruder (the Tanabe plastics company make), the heat set for 10 minutes was performed for the film in 100-degree C air oven. It was similarly estimated as the approach which performed the seal reinforcement of the obtained monolayer film, thermal resistance, and Hayes in the examples 1-5. A result is shown in Table 4. Heat-sealing reinforcement was not obtained.

[0122] (Example 8 of a comparison) The lactic-acid system polymer (B) of amorphism nature shown in Table 4 was used, extrusion membrane formation of the monolayer film with a thickness of 35 micrometers was carried out using the extruder (the Tanabe plastics company make), and the heat set for 10 minutes was performed for the film in 100-degree C air oven. It was similarly estimated as the approach which performed the obtained monolayer film thermal resistance, Hayes, and biodegradability in the examples 1-5. An evaluation result is shown in Table 4. About the thermal resistance of the laminated film produced here, the tear was seen with the test temperature of 60 degrees C and 80 degrees C in the real trial which used weight. The minimum value of the storage modulus (E') of a laminated film is 0 kgf/cm<sup>2</sup>, and there was no thermal resistance.

[0123]

[Table 4]

比較例		1	2	3	4	5	6	7	8
乳酸系ポリマー (A)	P1	P3	P2	P5	P4	P1	P2	—	—
融点 [℃]	163	168	176	162	162	163	176	—	—
結晶化温度 [℃]	110	108	110	73	93	110	110	—	—
乳酸系ポリマー (B)	P6	P8	P7	P9	P9	P7	—	P8	—
軟化温度 [℃]	52	46	46	57	57	52	—	46	—
厚み [μm]	35	35	35	35	50	35	35	35	35
ヘイズ [%]	5	7	1.0	1.1	9	13	15	10	—
耐熱性 実試験	60℃	○	○	○	○	×	×	○	×
	80℃	○	○	△	△	×	×	○	×
E' [kgf/cm²]	1000	1100	600	500	300	0	2600	0	—
生分解性	○	○	○	○	○	○	○	○	—
シール強度 [gf/15mm]	80℃	0	0	0	0	0	—	0	—
	90℃	0	0	0	0	0	—	0	—
	100℃	0	0	0	0	0	—	0	—
	120℃	0	0	0	0	0	—	0	—
	140℃	0	0	0	0	0	—	0	—

[0124]

[Effect of the Invention] This invention can provide various kinds of films, a sheet, a container, etc. with the lactic-acid system polymer layered product with the useful thermal resistance of 60 degrees C or more, the outstanding seal reinforcement, and biodegradability in which thermal melting arrival is possible.

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[Translation done.]

**\* NOTICES \***

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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

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**CLAIMS**

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**[Claim(s)]**

[Claim 1] The lactic-acid system polymer layered product which has the base material layer (I) which consists of a crystallized lactic-acid system polymer (A), and the sealing layer (II) which consists of a lactic-acid system polymer (B) of the amorphism nature which has softening temperature lower than the melting point of a lactic-acid system polymer (A) and in which thermal melting arrival is possible.

[Claim 2] The lactic-acid system polymer layered product according to claim 1 to which a lactic-acid system polymer (A) is characterized by being a lactic-acid system polymer with a melting point of 120 degrees C or more.

[Claim 3] The lactic-acid system polymer layered product according to claim 2 characterized by being the lactic-acid system polymer in which a lactic-acid system polymer (A) includes the polyester structural unit which carried out dehydration condensation of dicarboxylic acid and the diol, and/or the polyether structural unit which carried out dehydration condensation of dicarboxylic acid and the polyether polyol three to 60% of the weight.

[Claim 4] The lactic-acid system polymer layered product according to claim 2 or 3 to which a lactic-acid system polymer (A) is characterized by lactic-acid component content being 40% of the weight or more of a lactic-acid system polymer.

[Claim 5] The lactic-acid system polymer layered product according to claim 2 to which a lactic-acid system polymer (A) is characterized by being polylactic acid.

[Claim 6] The lactic-acid system polymer layered product of any one publication of claim 1-5 characterized by a lactic-acid system polymer (A) and/or a lactic-acid system polymer (B) consisting of the polymer to which deactivation of the polymerization catalyst was carried out by the quencher of a polymerization catalyst.

[Claim 7] The lactic-acid system polymer layered product of any one publication of claim 1-6 to which a lactic-acid system polymer (B) is characterized by being the lactic-acid system polymer of 40-110 degrees C of softening temperatures.

[Claim 8] The lactic-acid system polymer layered product of any one publication of claim 1-7 to which the base material layer (I) which consists of a lactic-acid system polymer (A) is characterized by crystallizing with a heat set.

[Claim 9] The lactic-acid system polymer layered product of any one publication of claim 1-8 to which the base material layer (I) which consists of a lactic-acid system polymer (A) is characterized by orientation being carried out by extension 1.4 to 16 times the field scale factor of this.

[Claim 10] The lactic-acid system polymer layered product of any one publication of claim 1-9 characterized by the minimum value of the storage modulus (E') below temperature lower 20 degrees C than the melting point being 400-40,000kg/cm<sup>2</sup> by the examining method (JIS- K-7198, A law) about the temperature dependence of dynamic viscoelasticity.

[Claim 11] The film using the lactic-acid system polymer layered product of any one publication of claim 1-10, or a sheet.

[Claim 12] The container using the lactic-acid system polymer layered product of any one publication of

claim 1-10.

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[Translation done.]